Journal of Cleaner Production 186 (2018) 883–893

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Integrated environmental management of pyrrhotite tailings at Raglan Mine: Part 2 desulphurized tailings as cover material

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ARTICLE INFO

Article history:
Available online 17 March 2018

Keywords:
Mine site reclamation
Covers with capillary barrier effects
Desulphurization
Acid mine drainage
Tailings reuse
Pyrrhotite

ABSTRACT

Mine site reclamation is a major concern for mining companies, especially when mine tailings are potentially acid generating mine drainage. For mine sites located in Arctic environments, many factors, such as climate change, the evolution of permafrost, remoteness, the availability of materials, and harsh climatic conditions, can make difficult the implementation of conventional rehabilitation methods. Therefore, it is necessary to design and develop reclamation approaches specific to these conditions. This study focuses on assessing the effectiveness of covers with capillary barrier effects, made of desulphurized tailings from Raglan mine (Nunavik region, Québec). The approach would mitigate the risk of water contamination by simultaneously limiting oxygen migration into potentially acid-generating tailings, as well as reducing their temperature. The study involved a detailed characterization of the cover materials and the construction of two instrumented columns in a controlled-environment chamber (two residual sulphide level). Column tests demonstrated that capillary barrier effects induced a permanent high degree of saturation (>85%) in the moisture-retaining layer made of desulphurized tailings. This high degree of saturation impeded oxygen migration; the estimated oxygen fluxes passing through the moisture-retaining layer being lower than 5.5×10^{-3} mol/m^2/day for the two columns. The column containing the desulphurized tailings with the highest sulphide content was slightly more efficient in controlling the oxygen flux due to the greater oxygen consumption by residual sulphides. Results also showed that freeze-thaw cycles did not significantly affect the hydrogeological properties of the desulphurized tailings, thus indicating a stability in the long-term performance of the cover.

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1. Introduction

Innovations in mining technologies now allow access to previously inaccessible ore deposits located in remote Arctic regions. Like any mines, these operations produce solid wastes, such as tailings and waste rocks that are stored at the surface in waste disposal areas. Such structures must be reclaimed at the end-life of the mine; but preferably during operation thorough progressive reclamation approach. The challenges related to reclamation are greater when mine wastes contain sulphide minerals which can oxidize and generate acid mine drainage (AMD). AMD is characterized by low pH and high concentrations of heavy metals, sulphates, and total dissolved solids (Lindsay et al., 2015); the AMD phenomenon can become harmful towards the environment. For northern mine sites, factors such as climate change, the presence of permafrost, remoteness, the availability of materials, and harsh climatic conditions can make the design and construction of reclamation scenarios even more difficult and costly than for mines in temperate climates (Bussière and Hayley, 2010).

Water and oxygen are essential reactants for AMD generation. Hence, most reclamation techniques aim to limit AMD formation by controlling the availability of one or both of these components

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https://doi.org/10.1016/j.jclepro.2018.03.132
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(MEND, 2001). In northern regions, AMD generation can also be limited by controlling the temperature of mine wastes through the addition of an insulation cover. Insulation covers function by shifting the active zone (i.e., the layer of permafrost that is subject to seasonal freezing and thawing) to within the cover. More specifically, the main objectives of this type of cover are: i) to promote freezing of the tailings which, in turn, help reduce contaminated water seepage to the environment, and ii) to slow the rate of sulphide oxidation and, consequently, AMD generation (e.g., MEND, 2012; Coulombe, 2012; Meldrum et al., 2001; Elberling, 2005; Elberling et al., 2000). However, progressive climate change is expected to result in the increasing thickness of the permafrost active zone (ACIA, 2004). Therefore, it is necessary to adapt reclamation methods implemented under such conditions to the effects of expected climate change in order to ensure long-term environmental protection.

From this perspective, one interesting alternative may consist on the use of multi-layered covers with capillary barrier effects (CCBEs) which, in northern conditions would have two main functions: i) maintain the tailings in a frozen/non-reactive state for the longest possible period, and ii) prevent oxygen fluxes from reaching potentially acid-generating tailings if they are unfrozen/reactive (Boulanger-Martel et al., 2015, 2016, 2017). Capillary barrier effects aim at maintaining one cover layer, which is commonly called the moisture-retaining layer (MRL), at a high degree of saturation (S_r) using the CCBE concept. Capillary barrier effects occur when a fine-grained material overlies a coarse-grained material. The difference in unsaturated hydraulic properties between the two contacting materials tends to restrict vertical water flow between the fine-grained and the coarse-grained layers, thus favouring a high degree of saturation in the fine-grained material (Nicholson et al., 1989; Aubertin et al., 1995, 1999; Bussière et al., 2003). A degree of saturation greater than 85–90% in the MRL was reported to significantly reduce the oxygen flux that can reach the base of a cover, thus controlling AMD generation in sulphidic tailings (e.g., Yanful, 1993; Aubertin et al., 1999; Mbonimpa et al., 2003).

This type of cover requires the use of a fine-grained (typically silty) material (Bussière et al., 2007). However, in northern environments, these materials are not always available close to the mine site. Thus, tailings desulphurization process becomes an attractive option because it can be used to transform potentially reactive tailings into cover materials for mine site reclamation (Benzaazoua et al., 2000; Bussière et al., 2004). The most adapted method to produce desulphurized tailings is non-selective froth flotation, which can be performed at the ore processing plant (Benzaazoua et al., 2000). This process reduces the proportion of problematic tailings by concentrating sulphides in a residual fraction (Benzaazoua et al., 2000, 2017). It also generates a desulphurized fraction that can be used as fine-grained material in a CCBE's moisture-retaining layer (Bussière et al., 2004). Several authors have confirmed the efficiency of CCBEs made of low-sulphide tailings in limiting oxygen diffusion (about 99%) and AMD generation through laboratory column tests (Aachib et al., 1994; Aubertin et al., 1995, 1997; Bussière et al., 1997, 1998, 2004; Kalonji-Kabambi et al., 2017) and field experimental cells (Bussière et al., 2007; Molson et al., 2008). In addition, the presence of residual sulphide minerals can have the advantage of reducing oxygen fluxes through a CCBE (at least in the short term) because they consume oxygen (Bussière et al., 2004; Demers et al., 2008). Ideally, sulphide recovery through the desulphurization process must reach a maximal level that guarantees that the material will not release any contaminants in the long term to the environment (Benzaazoua et al., 2017). In frozen or partially frozen conditions, O_2 diffusion, which is related to the effective oxygen diffusion coefficient (D_e), is greatly decreased by the presence of ice, which has a D_e (10^{-11} m^2/s) that is approximately two and six orders of magnitude lower than water (10^{-9} m^2/s) and air (10^{-5} m^2/s), respectively (e.g., Bortkovski, 2012; Fredlund et al., 2012; Elberling, 1998). The reactivity of tailings is also decreased with decreasing temperatures as described by the Arrhenius equation (e.g., Coulombe, 2012). Therefore, under such conditions, both the accessibility of oxygen and tailings' reactivity are reduced compared to the unfrozen state. This work will confirm whether or not the desulphurized tailings are efficient as a cover material in a CCBE to control AMD production from the Raglan tailings.

This study focuses on assessing the effectiveness of what is called hereinafter ICCBEs (insulation cover with capillary barrier effects, as proposed by Boulanger-Martel et al., 2017) with moisture-retaining layers made of desulphurized tailings in controlling AMD generation when exposed to cold climatic conditions (for details of desulphurized process see the companion article of Benzaazoua et al., 2017). The specific objectives of this study consist in: i) characterizing the hydrogeological and thermal properties of the tested desulphurized tailings; ii) assessing, by using column tests, the hydrogeological and thermal behaviour of ICCBE made of two sulphur levels of desulphurized tailing, both before and after freeze/thaw (F/T) cycles; and iii) evaluating the capacity of the ICCBEs to control oxygen migration, both before and after F/T cycles. The performance evaluation of the tested covers presented in this paper aims at evaluating the worst-case scenario where a complete thawing of the MRLs occurs and the tailings' reactivity reaches a maximum (i.e., at warmest temperature in the Raglan mine site). To the authors' knowledge, only two other laboratory research works assessed the CCBE technology for cold climatic conditions to control oxygen migration and the temperature of the mine wastes underneath. The first one tested crushed rock-bentonite mixtures as MRL materials in ICCBEs. The study showed that F/T cycles significantly affect the performance of the tested crushed rock-bentonite mixtures (Boulanger-Martel et al., 2015, 2016). The other study is ongoing and aims at testing fine compacted waste rock as MRL material (Boulanger-Martel et al., 2017). Another original aspect tackled in this study is the use of non-acid generating tailings in the construction of such ICCBEs, instead of natural soils. Repurposing tailings as construction materials for the reclamation of mine sites has been applied successfully in temperate climates (e.g., LTA mine site; Bussière et al., 2006), but this novel approach has not yet been tested under cold climatic conditions.

2. Materials and methods

2.1. Material preparation and characterization

To assess the possibility of using desulphurized tailings as a fine-grained material in the moisture-retaining layer of ICCBEs, a series of characterization tests were performed. The first step consisted of reducing the sulphide concentration in the acid-generating tailings through flotation. Non-selective sulphide flotation was carried-out on materials sampled at the end of the ore processing circuit at the Raglan Mine; i.e., tailings from the thickener output at a solids content of 60%. Water was added to the pulp to reach a solids content of 40%, which is similar to the solids content used during the flotation process at the mine. All flotation tests were performed using a Denver D-12 laboratory flotation machine with a cell volume of 2.5 L. The speed of the rotor-stator was adjusted to 1500 rpm and airflow was fixed at 10 l/min. The optimized pyrrhotite flotation conditions were: slurry pH = 10.5; copper sulfate ($CuSO_4$) activator concentration = 600 g/t; xanthate as collector

(KAX51) at a dosage of = 220 g/t; and methyl isobutyl carbinol (MIBC) frother was maintained constant at 13 μL in flotation cell. A depressant, carboxymethylcellulose (CMC), was used to eliminate the problem of floatable gangue minerals (e.g., serpentines, lizardite). More details on the environmental desulphurization process and the role of serpentine minerals can be found in Benzaazoua et al. (2017).

The main chemical, mineralogical, physical, hydrogeological, and thermal properties were determined for all materials used in the column tests; i.e., two desulphurized tailings with different residual sulphur and a crushed rock. In this study, most of crushed rock properties were obtained from characterization work performed by Coulombe (2012).

2.1.1. Physical and chemical characteristics

Characterization of the tested materials was performed using several analytical techniques. The elemental compositions of the desulphurized tailings were analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES; Perkin Elmer 3000 DV) and fusion whole rock by borate fused disc. Analysis of total inorganic carbon (C) and sulphur (S) concentrations were carried out using an induction furnace equipped with a dedicated infrared (IR) analyser (Eltra CS 2000) with a ±0.05 to 0.1 wt% precision (CEAEQ, 2013). Mineralogical phases were first determined by optical microscopy (OM), followed by both scanning electron microscopy (SEM; Hitachi S−3500 N) coupled with Energy Dispersive Spectrometry (EDS; Silicon Drift Detector X-Max 20 mm2 from Oxford) microanalysis, and X-ray diffraction (XRD; Bruker AXS Advance D8; XRD precision ± 1%; Bouzahzah, 2013). SEM microanalyses provided practical basic mineralogical mapping and an idea of elemental distribution within mineral phases. A reconciliation of the XRD mineralogical analysis (quantified by the Rietveld method) and chemistry results was performed (Bouzahzah, 2013; Taylor and Hinczak, 2001). The main chemical and mineralogical results are presented in Table 1 (for more details see Lessard, 2018; Benzaazoua et al., 2017).

The desulphurized tailings' residual sulphides (mainly pyrrhotite) were present mainly as inclusions in the gangue minerals (mostly in lizardite, which represents approximately 30% of the desulphurized tailings' mineralogical modal composition). Through environmental desulphurization, the pyrrhotite content of the tailings was lowered from 12.5% down to 0.64%. More than half of this pyrrhotite is included within lizardite and is much less reactive (Benzaazoua et al., 2017). Some amount of sulphide concentrate was added to the desulphurized tailings to prepare a second desulphurized tailings sample; more sulphidic with a pyrrhotite content of 1.55%, that would simulate a lower desulphurization process rate (less costly processes). Some traces of residual pentlandite and chalcopyrite were observed in both samples (about 0.29% and 0.40%, respectively). The main gangue minerals were actinolite (≈30%) and clinochlore (≈26%). Calcite (2.2%) is the only mineral in the Raglan desulphurized tailings that could have a significant acid-neutralizing potential. The main minerals identified in the crushed rock were actinolite (33%), albite (14%), anorthite (11%), and epidote (17%).

Static tests were performed on the desulphurized tailings to determine their neutralization potential (NP) and acid-generating potential (AP). The NP was evaluated using the Sobek et al. (1978) method as modified by Lawrence and Scheske (1997), while the AP was calculated based on the sulphur as sulphide content (in the sulphide form, $AP = S_{sulphide} \times 31.25$ kg $CaCO_3$/t). According to the net neutralization potential (NNP = NP-AP) and the NP/AP ratio criteria, which are frequently used to classify the AMD potential of mine wastes, the AMD-generating potential is uncertain for the two desulphurized tailings samples (the PNN and NPR values of desulphurized tailings #1 are respectively 13.56 and 1,94 and for desulphurized tailings #2, 2.84 and 1.11). The limits of the uncertainty zone are -20 kg $CaCO_3$/t < NNP <20 kg $CaCO_3$/t and 1 < NPR >2 (NPR values between 1.3 and 2.5 have a low risk of acid generation; Adam et al., 1997)). Normally, during the environmental assessment of a mining project, uncertain materials would need to undergo kinetic tests to specify the AMD potential (Benzaazoua et al., 2017).

The grain-size distribution of the desulphurized tailings was obtained using a Malvern Instruments Mastersizer S laser particle size analyser (see Fig. 1) (Lee Black et al., 1996). The grain-size distribution of the crushed rock was obtained by mechanical sieving (based on ASTM D422; ASTM, 2007) for particles larger than 300 μm and by laser diffraction for smaller grain sizes. According to the USCS soil classification (McCarthy, 2007), the desulphurized tailings are considered low plasticity silts (ML; similar to other hard

Table 1
Chemical and mineralogical properties.

Properties	Desulphurized tailings #1	Desulphurized tailings #2	Crushed rock (data from Coulombe, 2012)
S_{total} (%w/w)	0.46	0.81	0.24
C_{total} (%w/w)	0.26	0.26	0.32
AP (kg $CaCo_3$/t)	14.44	25.16	
NP (kg $CaCo_3$/t)	28		
NNP (kg $CaCo_3$/t)	13.56	2.84	
NRP (kg $CaCo_3$/t)	1.94	1.11	
Mineralogy (%w/w)			
Actinolite	29.75	30.00	33.6
Albite	2.76	2.78	14.4
Anorthite			11.2
Arsenopyrite	0.01	0.01	
Calcite	2.17	2.17	
Chalcopyrite	0.25	0.25	
Clinochlore	26.48	26.48	
Epidote			17.5
Lizardite	31.81	30.00	
Magnetite	2.56	2.00	
Orthoclase	1.37	1.30	
Pentlandite	0.29	0.40	
Pyrrhotite	0.64	1.55	
Quartz	1.60	2.78	8.3
Rutile	0.30	0.30	0.5

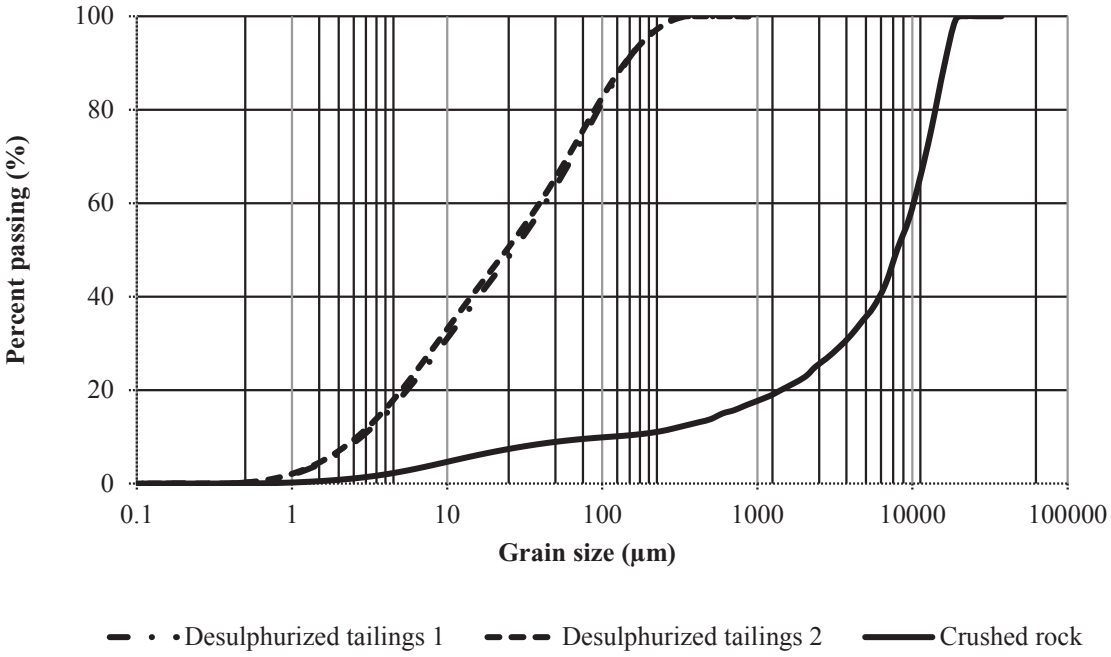

Fig. 1. Grain-size distributions of the tested materials (data for the crushed rock from Coulombe, 2012).

rock mine tailings; Bussière, 2007) and the crushed rock is classified as a well-graded gravel (GW). Based on the grain-size distribution, the contrast in properties between the fine and the coarse materials should be sufficient to allow for the creation of the desired capillary barrier effects; this was validated through determination the hydrogeological properties of the materials.

The determination of the specific gravity (G_s) was conducted in accordance with ASTM D854 (ASTM, 2014a) using a Micromeritics Accupyc 1330 pycnometer (±0.01% accuracy). The G_s values are similar for the two desulphurized tailings (2.86), and higher (3.01) for the crushed rock (Table 2) because the molecular weight of epidote.

2.1.2. Hydrogeological and thermal properties

Since the two desulphurized tailings samples have very similar physical properties, hydrogeological characterization was performed only on desulphurized tailings #1. Permeability tests, to evaluate the materials' saturated hydraulic conductivities (k_{sat}), were conducted in rigid wall permeameters following ASTM D5856 (ASTM, 2015) for the falling head and ASTM D2434-68 (ASTM, 2006) for the constant head test methods for desulphurized tailings #1 and the crushed rock, respectively. The measured k_{sat} values were 1.1×10^{-5} cm/s for the desulphurized tailings and 3.4×10^{-1} cm/s for the crushed rock. This difference in k_{sat} values confirms the significant hydrogeological contrast between the two materials. Since the physical models (experimental columns) were subjected to freeze/thaw cycles, the k_{sat} value for the desulphurized tailings #1 was also determined after eight F/T cycles. To do so, the same sample used to measure the initial k_{sat} value was kept in its permeameter and submitted to F/T cycles. The minimum temperature of −20 °C was reached in a conventional freezer, while in the thawed state, the sample was warmed to room temperature (20 °C). To simulate downward freezing, the side and bottom of the permeameter were insulated with polystyrene. Temperatures were monitored with a Decagon RT-1 probe positioned between the permeameter and the insulation layer to control the temperature at −13 °C. The k_{sat} value measured after eight F/T cycles was 7.2×10^{-5} cm/s. This indicates that F/T cycles have no significant impact on the saturated hydraulic conductivity of the desulphurized tailings.

Table 2
Physical, geotechnical, chemical, and thermal properties of the tested cover materials.

Properties	Desulphurized tailings #1	Desulphurized tailings #2	Crushed rock (data from Coulombe, 2012)
G_s (−)	2.85		3.01
k_{sat} (cm/sec)	1.1×10^{-5}		3.4×10^{-1}
$k_{sat\ after\ 8\ F/T\ cycles}$ (cm/sec)	7.2×10^{-5}		–
AEV (cm)	160		5
ψ_r (cm)	10 020		70
θ_s (−)	0.38		0.38
θ_r (−)	0.03		0.02
$\lambda_{u\ Sr\ 94–98\%}$ (W/m°C)	1.55	1.24	–
$\lambda_{f\ Sr\ 94–98\%}$ (W/m°C)	2.30	2.02	–
$\lambda_{u\ Sr\ 27\%}$ (W/m°C)	–	–	1.54
$\lambda_{f\ Sr\ 27\%}$ (W/m°C)	–	–	1.65
λ_s (W/m°C)	3.39	3.42	3.26
K_r (day)	1.12×10^{-2}	1.19×10^{-2}	–

The drainage water retention curve (WRC) of the desulphurized tailings #1 was obtained with a Tempe cell following ASTM D6836 (ASTM, 2016). Important parameters describing material's hydro-geological behaviour, including: the air entry value (AEV or ψ_a), residual suction (ψ_r), volumetric water content at saturation (θ_s), and residual volumetric water content (θ_r), have been estimated from the WRC and are presented in Table 2. Such values were obtained for desulphurized tailings #1 using the tangent method (Fredlund et al., 2012). For the crushed rocks, a drainage column was used by Coulombe (2012) to obtain the WRC. This method is more appropriate for such coarse-grained materials (Chapuis et al., 2007). The column that was used was 50 cm in height and 14 cm in diameter, and equipped with tensiometers and 5TM Decagon probes. The column was saturated before the test; suction and volumetric water content measurements were taken simultaneously at two locations in the column during the drainage phase to obtain the WRC (for more details see Coulombe, 2012). Tests were performed at a sample porosity of 0.38 for both the crushed rock and the desulphurized tailings. The results presented in Table 2 show that the tailings have good water retention properties compared to the crushed rock (AEV between 160 and 300 cm of water for the tailings, compared to 5 cm of water for the crushed rock). For an efficient CCBE, the AEV of the fine-grained material should not be reached before the coarse-grained material reaches its ψ_r (Bussière et al., 2003). It is also suggested that the difference between the k_{sat} of the fine- and coarse-grained material should be more than three orders of magnitude (Aubertin et al., 1995), which is the case for these materials. As shown in Table 2, the contrasting properties of the fine and coarse-grained materials should allow for the creation of the capillary barrier effects needed for the CCBE.

A Hukseflux thermal sensor TP08 needle probe was used to determine the unfrozen (λ_u) and frozen (λ_f) thermal conductivities of the desulphurized tailings; these tests were based on ASTM D5334 (ASTM, 2014b). The needle probe test method is recommended for granular soils with a particle diameter lower than 0.5 mm and has been used by others to determine the thermal conductivity of tailings (e.g., Coulombe, 2012; Nixon, 2000). Thermal conductivity measurements were made near saturation in the unfrozen and frozen states for desulphurized tailings #1 and #2. Tests were conducted on samples having total porosities of 0.43, and degrees of saturation of 94 and 98%, respectively. The testing temperatures were of 5 ± 1 °C for the unfrozen state and -5 ± 1 °C for the frozen state. For desulphurized tailings #1 and #2, the λ_u were 1.55 and 1.24 W/m °C, respectively, and the λ_f were 2.30 and 2.02 W/m °C, respectively. The obtained thermal conductivities are summarized in Table 2 and are typical for this type of fine-grained material (Côté and Konrad, 2005; Fredlund et al., 2012). The thermal conductivity of solid particles (λ_s) was also calculated based on the tailings' mineralogical composition and the thermal conductivity of rock-forming minerals using the geometric mean approach suggested by Côté and Konrad (2005). The calculated thermal conductivities of the solid particles of desulphurized tailings #1 and #2 were of 3.39 and 3.42 W/m°C, respectively (refer to Table 2).

A detailed characterization of the thermal properties was also performed by Coulombe (2012) for the crushed rock. The thermal conductivity of the crushed rock was measured at various degrees of saturation (between 4 and 57%), and in both the unfrozen (average temperature of 5 °C) and frozen (average temperature of −5 °C) states using a heat fluxmeter (Côté and Konrad, 2005). In this study, the typical degree of saturation for the crushed rock was near 30%. Therefore, the unfrozen (1.54 W/m °C) and frozen (1.65 W/m °C) thermal conductivities measured at a degree of saturation of 27% by Coulombe (2012) are presented in Table 2 as reference values. Based on the geometric mean approach, an average thermal conductivity of solid particles in the crushed rock was estimated at 3.26 W/m °C by Coulombe (2012).

2.1.3. Reactivity of the desulphurized tailings

In addition to the effective diffusion coefficient (D_e), the sulphide reaction rate coefficient (K_r) is an essential parameter to evaluate the migration of oxygen through the tested CCBE since the desulphurized tailings contain a residual concentration of sulphide minerals that will consume part of the diffusive oxygen that migrates through the cover. The K_r of the desulphurized tailings (#1 and #2) was measured with a two-chamber diffusion/consumption cell based on the technique developed by Mbonimpa et al. (2003). A sample of the tested material is placed on a perforate plate in a sealed chamber at a targeted degree of saturation of ~50% and a porosity of 0.43. This S_r was selected based on the results of Coulombe (2012) who demonstrated that the oxidation of the Raglan tailings was optimal when the S_r was between 40% and 60%. Before the test, oxygen is purged from the cell, using nitrogen gas, until the oxygen concentrations in the two chambers becomes nil. An Apogee SO-110 probe, which has a sensitivity of 1% O_2, was used to monitor oxygen concentrations in both the top and bottom chambers. At the start of the test, the top chamber is temporarily opened to introduce atmospheric oxygen. The oxygen concentration is then measured in both chambers until it reaches a similar value in each. For desulphurized tailings #1 and #2, the test duration was typically around four days.

The POLLUTEv6 numerical code was used to simulate oxygen migration into the diffusion/consumption cell and to determine the first order sulphide reaction rate coefficient parameter (for more details on the numerical code see Rowe et al., 1994). This software has been used by others to determine the oxygen diffusion coefficient and oxygen flux through various non-reactive and reactive materials (e.g., Yanful, 1993; Aubertin et al., 1995, 2000; Mbonimpa et al., 2003, 2011; Aachib et al., 2004). To perform the numerical analysis, the following parameters must be entered into the code: the porosity ($n = \theta_{eq}$ = equivalent porosity), Darcy's velocity ($v = 0$), the half-life degradation $t^*_{1/2}$, and the coefficient of hydrodynamic dispersion ($D^* = D_e\theta_{eq}^{-1}$). The equivalent porosity (Equation (1)) is a parameter that was introduced to account for the oxygen transport that occurs in both air (θ_a, the volumetric air content) and water components (θ_w, the volumetric water content; Aubertin et al., 1999, 2000):

$$\theta_{eq} = \theta_a + H\theta_w \quad [1]$$

where H is the dimensionless Henry's equilibrium constant (0.03 at 20 °C). The D_e parameter was calculated with the Aachib et al. (2004) model. The model is a semi-empirical expression developed to estimate D_e based on the material's properties, such as its porosity and the volumetric air and water contents (Aachib et al., 2004):

$$D_e = 1 \Big/ n^2 \left(D_a^0 \theta_a^{p_a} + H D_w^0 \theta_w^{p_w} \right) \quad [2]$$

where D_a^0 and D_W^0 are the O_2 diffusion coefficients in air and water, respectively; and p_a and p_w are related to the tortuosity of the gaseous and liquid interstitial phases, respectively. As suggested by Aachib et al. (2004), a value of $p_a = p_w = 3.4$ was used in this study. In POLLUTEv6, the reactivity is represented through a half-life degradation $t^*_{1/2}$ (Mbonimpa et al., 2003):

$$t_{1/2}^* = \ln 2 / K_r^* = \theta_{eq} \frac{\ln 2}{K_r} \quad [3]$$

where $K_r^*(=K_r/\theta_{eq})$ is the bulk first order sulphide reaction rate

coefficient. A boundary condition of finite mass initial source (20.9% of O_2) was set at the top of the cell and a boundary condition of fixed outflow (n = 1) was imposed at the bottom. Material characteristics and the conceptual numerical model are summarized in Fig. 2. Once all parameters were entered into the numerical model, the K_r (or $t^*_{1/2}$) was modified until the observed O_2 concentrations fit with the POLLUTE predictions.

The obtained K_r values are presented in Table 2 and are 1.2×10^{-2} and 1.9×10^{-2} day^{-1} for desulphurized tailings #1 and #2, respectively. The oxygen diffusion/consumption test results for both desulphurized tailings show a typical behaviour for a low-reactivity material suitable for reuse as cover material (Demers et al., 2009; Gosselin, 2007).

2.2. Columns design

Column tests were performed to assess the performance of CCBEs made with desulphurized tailings in controlling oxygen migration under cold climatic conditions. The main features of these columns are presented below.

2.2.1. Configuration and instrumentation

The configurations of the instrumented columns used for this study is shown in Fig. 3. Two columns (2.1 m high columns with and 0.14 m inner diameter) were built with different CCBE variants. From bottom to top, the CCBEs were made of a coarse-grained support layer (to generate capillary barrier effects), a 0.7 m thick MRL, and a 0.4 m thick protective layer. The support and protective layers were made of crushed rock while the MRLs were made with desulphurized tailings. The MRLs in columns 1 (C1) and 2 (C2) were made of desulphurized tailings #1 and #2, respectively. For the crushed rock, compaction was performed to reach a porosity of 0.25, which corresponds to a dry density (ρ_d) of 2260 kg/m^3. For the desulphurized tailings, compaction yielded a porosity between 0.36 and 0.40. The initial volumetric water content was 0.36 (S_r between 90 and 100%) for both desulphurized tailings. To facilitate setup, the columns were filled in layers of about 1.5 kg (corresponding to approximately a 0.1 m lift thickness) that were compacted to meet the targeted porosities.

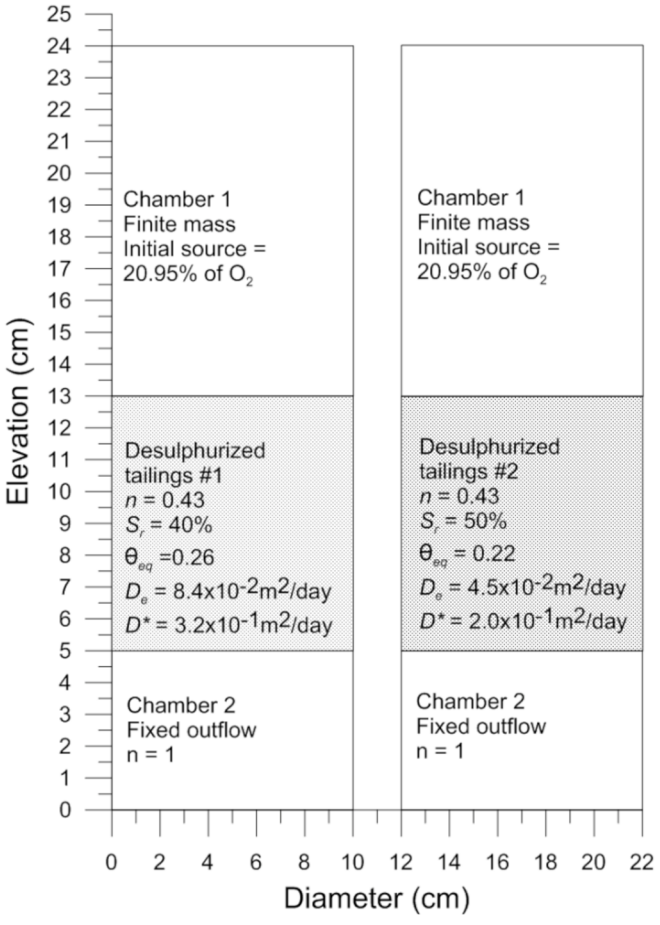

Fig. 2. Conceptual numerical model used to simulate oxygen diffusion/consumption tests.

The columns were made of 7/8 ″ thick high-density polyethylene (HDPE). Due to their height, C1 and C2 were segmented into two separated and fixable parts to facilitate instrumentation installation and filling. The joint between the two column sections was located above the capillary break, at the bottom of the MRL in order to avoid any disturbance (see Fig. 3). Vitaulic® joints were used for this junction and at the base of the columns. The columns' bases were custom-made polyethylene supports designed for receiving a porous ceramic plate. These bases were also equipped with tube fittings to allow water drainage and saturation of the ceramic plate. Gas purge valves were installed in the bottom support.

The columns were instrumented at different heights with seven Decagon 5TM temperature and volumetric water content sensors (Fig. 3). Decagon 5TM probes use the frequency domain (FD) technique (e.g., Malicki, 1983; Robinson and Dean, 1993; Hilhorst and Dirksen, 1994) to estimate θ_w. When priory calibrated with a specific material, the probe has an accuracy of ±0.03 m^3/m^3 for a range of 0–100% humidity. The Em50 data logger automatically interprets the electrical signals sent by the probes to yield a volumetric water content value according to the manufacturer's calibration; the values were corrected with a specific calibration equation for each material (for details see Lessard, 2018). A Decagon MPS-1 probe was also installed in the center of the MRL for matric suction measurements (results not presented here; see Lessard, 2018). Apogee SO-110 sensors were used to log the O_2 concentrations inside the bottom capillary break layer, at 5 cm below the MRL (Fig. 3). Additionally, gas sampling ports (see Demers et al., 2009) were added to measure O_2 concentrations at various heights in the columns; these measurements were used to calibrate the O_2 sensors (Boulanger-Martel et al., 2015) and to calculate the oxygen flux using the oxygen gradient method (Demers et al., 2009). To create more realistic conditions, the columns were insulated with 25.4 cm of Styrofoam to favour unidimensional freezing and thawing.

2.2.2. Experimental approach

The goal of the column tests was to evaluate the ability of the two ICCBEs (made with MRLs from different desulphurized tailings) to control oxygen migration, as well as to determine the influence of F/T cycles on the ICCBEs' performances. The parameters studied were: the O_2 concentration in the MRLs, the evolution of temperatures, and the distribution of θ_w in the tested ICCBEs. A typical testing sequence is illustrated in Fig. 4. The columns were first exposed to ambient temperature for a period of 27 days. A 7-day infiltration test was then performed at room temperature with two additions of 1.6 L of water at the top of each column. Rinse water quality (pH, Eh, conductivity, acidity, and alkalinity) was measured for each column (presented in Lessard, 2018). Next, the columns were exposed to ambient temperature for a 10-day period. Afterwards, a 20-day F/T cycle was applied to the columns; i.e., 10 days of freezing and 10 days of thawing. The tests were carried out in a specially designed proportional-integral-derivative (PID)

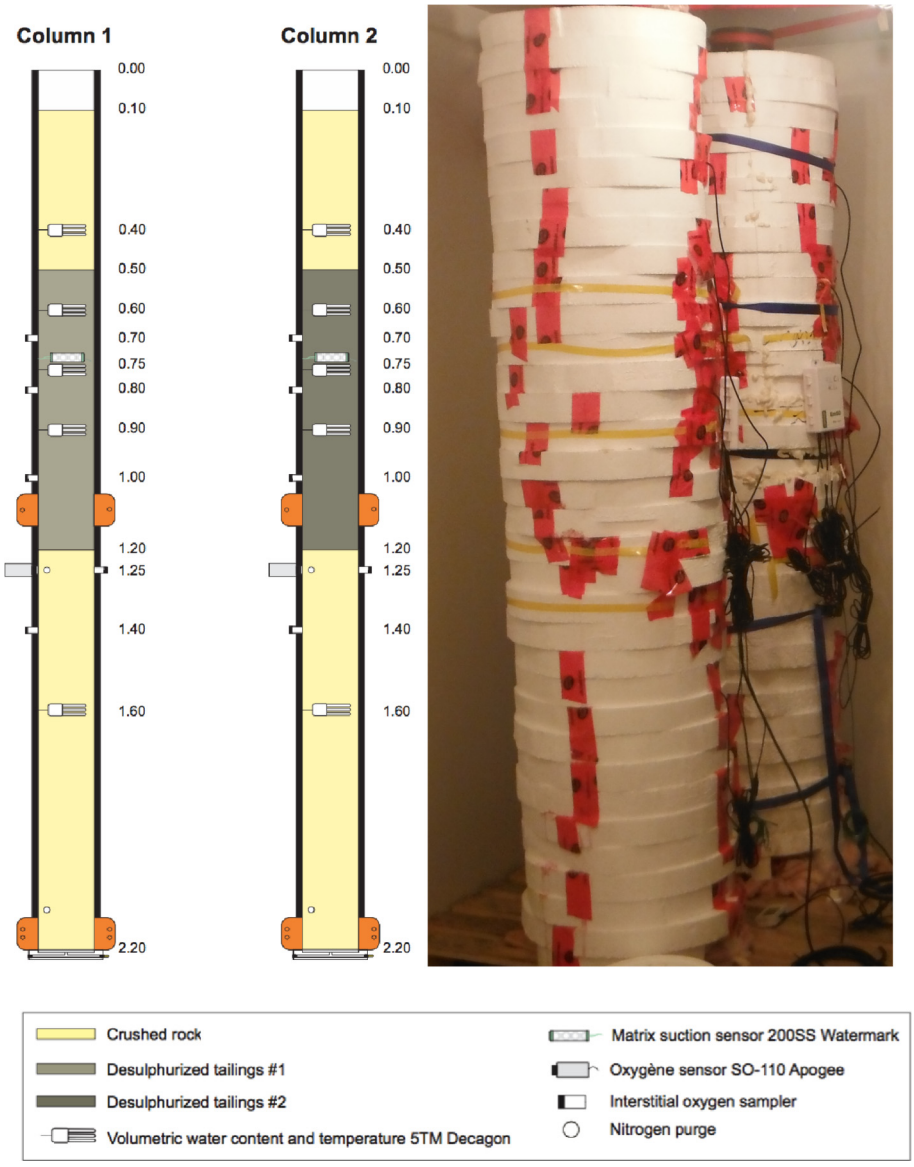

Fig. 3. Column configurations and instrumentation (left) and a photo of the columns in the controlled-atmosphere chamber (right).

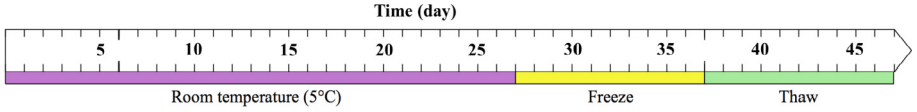

Fig. 4. Typical column testing cycle.

controlled industrial freezer at temperatures characteristic of those at the Raglan mine site; temperatures were set at 5 °C for the thawing cycles and at −13 °C for the freezing cycles. Similar testing conditions were used by Boulanger-Martel et al. (2015, 2016). In this study, the columns were subjected to eight complete F/T and wetting-drainage cycles over a period of about a year. Oxygen concentration measurements were not performed at each cycle. Oxygen concentration measurements were taken each day for 12 days after the last thaw cycle. Similar O_2 concentration gradients were measured from days 6–12. Therefore, representative O_2 concentration measurements were used for oxygen flux calculations.

3. Results

This section discusses the column test results pertaining to the relevance of using desulphurized tailings as cover materials under cold climatic conditions.

3.1. Hydrogeological behaviour

The AEVs of the desulphurized tailings MRLs were higher (AEV = 16 kPa) than the values obtained for the coarser underlying material (crushed rock AEV = 0.5 kPa). This cover configuration should theoretically induce capillary barrier effects and create a

barrier to vertical water flow, thus promoting water retention in the MRL. Typically, the volumetric water content profile for a given CCBE involves the two crushed rock layers containing close to their residual volumetric water contents, while the MRL remains at a high volumetric water content (ideally, corresponding to an $S_r > 85\%$; Aubertin et al., 1995, 1997; Bussière et al., 2007).

Fig. 5 show the profiles of temperature (T) and θ_w versus time (t) obtained for C1 and C2, respectively, for eight F/T cycles. Based on the measurements taken at different elevations in the columns, the values of θ_w can be considered homogeneous throughout the MRL for the frozen ($\theta_w \approx 0.15$) and unfrozen ($\theta_w \approx 0.35$) states. For the upper layer of crushed rock, the average values of θ_w for the frozen and unfrozen states are $\theta_w = 0.04$ and $\theta_w = 0.06$, respectively. For the bottom capillary break layer, also made of crushed rock, the average values of θ_w for the frozen and unfrozen states are $\theta_w = 0.07$ and $\theta_w = 0.10$, respectively. These results show that the hydrogeological behaviour (volumetric water content profile) is typical of an efficient CCBE where high volumetric water contents are observed in the MRL and close to residual volumetric water contents are observed in the two coarse-grained layers. The obtained unfrozen θ_w values were similar to the targeted values ($0.35 < \theta_w < 0.37$ for $0.36 < n < 0.40$) that would allow for an $S_r > 85\%$. Additionally, results showed that F/T cycles did not significantly affect the hydrogeological behaviour of the ICCBEs after eight cycles because the observed θ_w values were similar for each cycle. This confirms that the capacity of desulphurized tailings to regain their hydrogeological properties in the course of F/T cycles, a phenomenon comparable to that of non-plastic soils, such as silts (Bussière et al., 2007).

3.2. Thermal behaviour

The temperature evolution in the various column layers shows that when the temperature of the controlled-atmosphere chamber changed (to 10 °C for thawing and −18 °C for freezing), there was a relatively rapid freezing or thawing of the interstitial water until the material reached approximately 0 °C (see sensors at depths of 0.60, 0.75, and 0.90 m in Fig. 3). At this temperature (0 °C), there is release of latent heat during the phase transition, which explains the plateau in θ_w vs. time for the MRL. This discontinuity is called the 'zero curtain effect' and occurs at the frozen front in most partially saturated soils (e.g., Harlan and Nixon, 1978; Outcalt et al., 1990). Due to high θ_w values in the MRL, the solidification and fusion consume energy, which delays heat transfer. Because of the lower volumetric water contents recorded in the two coarse-grained layers (see sensors at depths of 0.40 and 1.60 m in Fig. 3), there was no plateau during the F/T process. Furthermore, the evolutions of T and unfrozen θ_w were in good agreement. During freezing, unfrozen θ_w dropped until T reached 0 °C. When most of the water became ice, T started to drop again to values close to the value in the controlled-atmosphere chamber. A similar behaviour was observed during thawing with a progressive increase in the unfrozen θ_w. When most of the ice melted and the unfrozen θ_w became stable in the MRL, the temperature started to increase to the value in the controlled-atmosphere chamber.

3.3. Oxygen flux calculations

The goal of using ICCBEs in cold climatic conditions is to reduce the time that tailings spend above the freezing temperature and to control oxygen fluxes when the tailings are reactive and unfrozen. As mentioned earlier, this study evaluated a worst-case scenario where the MRLs were completely thawed. Therefore, oxygen flux calculations were conducted for the conditions observed after thawing.

Three approaches to assess the vertical oxygen flux migrating through the ICCBEs were used in this study. The first and simplest approach uses the unidimensional Fick's first law for a non-reactive media (Mbonimpa et al., 2003):

$$F_{s,L} = C_0 D_e / L \quad [4]$$

In this equation, the $F_{s,L}$ parameter is the steady-state vertical oxygen flux at the base of a non-reactive cover layer material, assuming that all oxygen is consumed at the base of the cover. C_0 is the concentration of oxygen in the atmosphere; L is the MRL's thickness (it is assumed here in that the two capillary break layers don't influence the oxygen diffusion due to their low volumetric

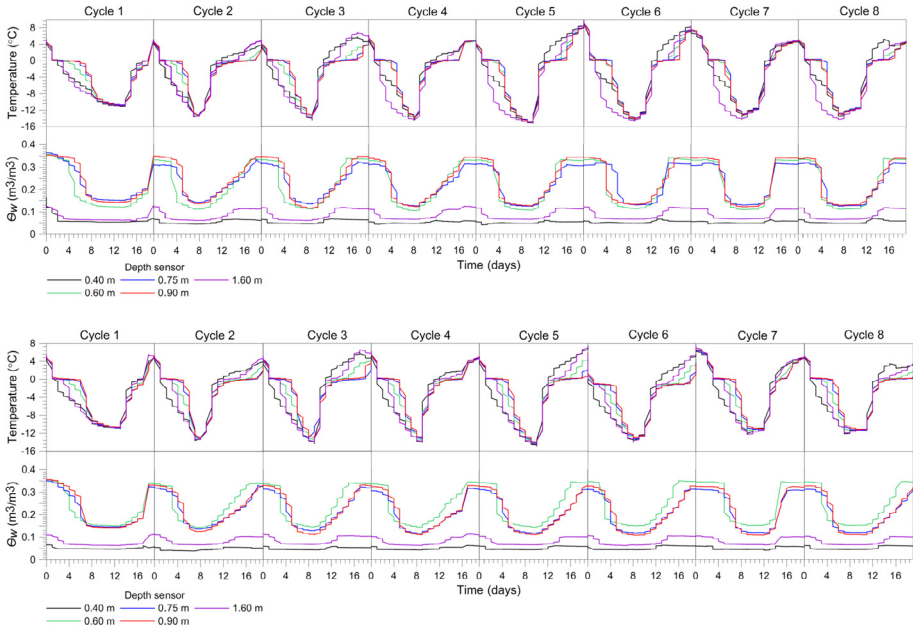

Fig. 5. Evolution of temperatures and unfrozen volumetric water contents (θ_w) over eight F/T cycles in Column 1 and 2.

water contents; Mbonimpa et al., 2003), and D_e is the effective diffusion coefficient estimated using the volumetric water content measurements and Equation (2).

The second approach, developed by Mbonimpa et al. (2003), is an equation to assess the steady-state diffusive oxygen flux ($F_{sR,L}$) below the oxygen-barrier layer (at $z = L$) of a reactive MRL with a thickness of L (Equation (5)). Again, it is assumed that there is a rapid and complete consumption of O_2 below the cover, and that θ_w and D_e are homogeneous throughout the cover materials. The K_r^* parameter is needed for calculating oxygen fluxes through a reactive cover. The K_r^* values measured in both desulphurized tailings samples (Table 2) using the diffusion/consumption cell tests were used for porosity values of 0.36–0.40 (Table 2). The initial oxygen concentration in the reactive material is assumed to be 0.

$$F_{sR,L} = F_{s,L} + 2K_r^* F_{s,L} \sum_{i=1}^{\infty} \left((-1)^i \Big/ \left(i^2 \pi^2 D^* \Big/ L^2 + k_r^* \right) \right) \quad [5]$$

In the third approach the vertical oxygen flux can be estimated using the oxygen gradient method (Elberling et al., 1993, 1994; Demers et al., 2009). Diffusion of gas through a soil cover (reactive or non-reactive) may be estimated using Fick's first law, where the flux is equal to the effective diffusion coefficient, D_e, multiplied by the concentration gradient.

$$F_{s,L} = -D_e \partial c / \partial z \quad [6]$$

For flux calculations, the concentration gradient is obtained using the oxygen concentrations measured at two gas sampling points in the MRL at a given time, and D_e (2.1×10^{-4} m^2/day) is estimated from volumetric water content measurements ($\theta_w = 0.32$) and Equation (2). Gas samples were taken once per day over 12 day at elevations of 0.80 and 1 m in both columns (Fig. 3). A representative oxygen concentration profile was achieved on the sixth day and the values measured on that day were used for further flux calculations (C1: 0.80 m = 5.3 mol O_2/m^3 and 1 m = 4.5 mol O_2/m^3; C2: 0.80 m = 3.2 mol O_2/m^3 and 1 m = 2.1 mol O_2/m^3).

The D_e parameter is key to estimating the O_2 flux in all three approaches. As previously discussed, one of the key parameters to estimate D_e is the degree of saturation ($S_r = \theta_w/n$). In the O_2 flux calculations, a range of realistic values were used (S_r between 87.5% and 97%) based on potential porosities in the tested columns. All results of O_2 flux obtained with Fick's law for non-reactive media and the methods suggested by Mbonimpa et al. (2003) are presented in Table 3. For the approach using Equation (4) (Fick's law for non-reactive materials), the O_2 flux was estimated to be between 2.1×10^{-3} to 1.9×10^{-5} mol/m^2/day. When the reactivity of the MRL is integrated into the calculations (using the equation suggested by Mbonimpa et al., 2003), the O_2 flux ranged from 1.1×10^{-4} to 6.4×10^{-9} mol/m^2/day and from 1.7×10^{-5} to 1.3×10^{-9} mol/m^2/day for C1 and C2, respectively. Using the oxygen gradient method, the calculated O_2 fluxes ranged from 7.7×10^{-3} to 9.1×10^{-4} mol/m^2/day and from 9.5×10^{-3} to 1.1×10^{-3} mol/m^2/day for C1 and C2, respectively. For these calculations, the parameter D_e was estimated using Equation (2), the θ_w was measured the same day that the oxygen concentration profile is considered representative (day 6) and for porosity values between 0.36 and 0.40.

Typically, for an effective CCBE, the targeted O_2 flux is less than 5.5×10^{-3} mol/m^2/day (e.g., Bussière, 1999; Dagenais, 2005; Nastev and Aubertin, 2000), these results show that the tested ICCBEs usually meet this criterion. As predicted with the equation suggested by Mbonimpa et al. (2003), the oxygen fluxes passing through the ICCBE in C2 were slightly lower than for the ICCBE in C1 due to the slightly higher reactivity of desulphurized tailings #2. The fluxes measured with the oxygen gradient method were similar to those theoretically predicted based on Equation (4) (Fick's law for non-reactive media). In this context, the steady-state fluxes with the non-reactive method were conservative results relative to the results of oxygen gradient method and the Mbonimpa et al. (2003) approach (Equation (5)), which accounts for O_2 consumption by the MRL. However, the action of residual sulphide reactivity (contained in the MRL) on O_2 diffusion decreases with time. Which means that the O_2 fluxes predicted using the equation suggested by Mbonimpa et al. (2003) will increase with time to reach values similar to those given by the two other methods.

4. Conclusions

In arctic regions, one of the most commonly recommended reclamation methods for tailings storage facilities are insulating covers, which aim to keep tailings permanently frozen and non-reactive (MEND, 2004; MEND, 2009; MEND, 2012). However, this method of controlling AMD generation could be sensitive to increases in the thickness of the permafrost's active zone that are anticipated due to climate change. In order to improve the robustness of mine site reclamation covers in arctic regions, the CCBE concept can be combined with an insulation objective to create new type of cover that would act as both, an oxygen barrier during the summer, and a thermal barrier during the rest of the year. Desulphurized tailings represent an interesting alternative to fine-grained cover material because of their on-site availability and their advantageous hydrogeological properties. In this study, two ICCBEs with MRLs made from two desulphurized levels tailings were tested in columns in a controlled-atmosphere chamber to assess their performance when exposed to conditions similar to those found in the Arctic.

Characterization of the cover materials showed that they have adequate physical and hydrogeological properties to generate the

Table 3
Material characteristics and oxygen flux results.

Material	n (−)	S_r (%)	θ_a (−)	θ_{eq} (−)	D_e (m^2/day)	$F_{s,L}$ (mol/m^2/day)	$F_{s,R,L}$ (mol/m^2/day)
Desulphurized tailings #1	0.36	100.0	0.00	0.01	1.6×10^{-6}	1.9×10^{-5}	6.4×10^{-9}
	0.37	97.3	0.01	0.02	3.3×10^{-6}	4.0×10^{-5}	6.4×10^{-9}
	0.38	94.7	0.02	0.03	1.9×10^{-5}	2.4×10^{-4}	1.6×10^{-9}
	0.39	92.3	0.03	0.04	6.9×10^{-5}	8.5×10^{-4}	3.2×10^{-6}
	0.40	90.0	0.04	0.05	1.7×10^{-4}	2.1×10^{-3}	1.1×10^{-4}
Desulphurized tailings #2	0.36	100.0	0.00	0.02	1.6×10^{-6}	4.1×10^{-5}	1.3×10^{-9}
	0.37	97.3	0.01	0.03	3.3×10^{-6}	2.5×10^{-4}	1.3×10^{-9}
	0.38	94.7	0.02	0.04	1.9×10^{-5}	9.0×10^{-4}	1.3×10^{-9}
	0.39	92.3	0.03	0.05	6.9×10^{-5}	2.2×10^{-3}	1.4×10^{-7}
	0.40	90.0	0.04	0.06	1.7×10^{-4}	4.5×10^{-3}	1.7×10^{-5}

capillary barrier effects necessary for a CCBEs. Results from the two-chamber diffusion/consumption cell tests allowed to obtain estimated K_r values of the desulphurized tailings samples. Volumetric water content measurements in both tested ICCBEs confirm the presence of the capillary barrier effects with a MRL having a high degree of saturation (>85%) while the two coarse-grained layers are mostly desaturated. Freeze/thaw cycles did not significantly influence the hydrogeological behaviour of the tested ICCBEs, at least over the eight F/T cycles performed in this study.

Oxygen flux migration though the ICCBEs was evaluated with three different approaches: i) Fick's law for non-reactive media; ii) Mbonimpa et al. (2003) equation for reactive material; and iii) the oxygen gradient method. The oxygen fluxes obtained with these three approaches are in good agreement and are lower than the usual maximum desired flux of 5.5×10^{-3} mol/m^2/day. The Mbonimpa et al. (2003) equation showed that sulphides in the MRL reduced (in the short term) oxygen fluxes ($<1.11 \times 10^{-4}$ mol/m^2/day) that migrated through the MRL. These results proved that the CCBE configuration presented in this study could be efficient in controlling O_2 migration from the atmosphere towards covered Raglan mine tailings. It appears that the column that had the highest residual sulphide content reduced the oxygen flux more effectively due to O_2 consumption in the MRL. However, there must be a balance between the O_2 consumption capacity of the MRL material and its contaminant-generation potential. Hence, in the case of desulphurized tailings #2 used in this study, further study is needed to ensure the material is not generating contaminants (NNP = 2.8 kg $CaCO_3$/t and NPR = 1.1).

Acknowledgements

This study was funded by the Industrial NSERC Industrial Chair on Mine Site Reclamation and by the Research Institute on Mines and the Environment (RIME UQAT-Polytechnique; www.irme.ca). The authors wish to thank the staff at Raglan mine's environmental department (Mélanie Côté and Louis Marcoux) for their support, as well as the personnel at URSTM (in particular Mathieu Villeneuve, Hassan Bouzahzah, Alain Perreault, Patrick Bernèche, Joel Beauregard, and Yvan Poirier) for their assistance during the laboratory work.

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